MISTE: MICROGRAVITY EXPERIMENT TO MEASURE THE HEAT CAPACITY AND SUSCEPTIBILITY NEAR A LIQUID-GAS CRITICAL POINT

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Abstract

An experiment called MISTE (Microgravity Scaling Theory Experiment) is being developed for a future International Space Station mission. MISTE will be performed in the Low Temperature Microgravity Physics Experiments Facility located on the Japanese Experimental Module – Exposed Facility. The goal of this experiment is to provide a stringent test of scaling theory predictions for critical phenomena near a liquid-gas critical point both in the asymptotic and crossover regions. This experiment is designed to perform PVT, heat capacity at constant volume, and isothermal susceptibility measurements near the ^{3}He liquid-gas critical point. Measurements will be performed throughout the critical region with particular emphasis along the critical isochore, critical isotherm, and coexistence curve. In recent years, values for the critical exponents and universal asymptotic amplitude ratios have been improved using Renormalization Group theory. Combining asymptotic measurements with those obtained in the crossover region farther away from the transition will also provide an important test of crossover theories. This planned microgravity study will require high-resolution pressure, density, and temperature sensors that operate at liquid helium temperatures. A description of the experimental approach for performing these critical point measurements is discussed and results of ground-based measurements in preparation for the flight experiment are presented.

Introduction

The last several decades have witnessed widespread efforts to test theoretical predictions near critical points. In the early years emphasis was placed on theoretical and experimental studies of the liquid-gas critical point because of the fact that this system permitted the

measurement of a wide variety of thermophysical properties within the critical region. Unfortunately, experimental studies of the liquid-gas critical point are subject to limitations resulting from the effects of gravity.[1] In this system, gravity couples directly to the order parameter, which is the difference between the system's density and the critical density. In ground-based laboratories, a gravity induced vertical density stratification does not permit accurate thermodynamic measurements in the asymptotic region close the critical point. In the case of the ^{3}He liquid-gas critical point, ground-based measurements[2] along a path of constant critical density, $\rho = \rho_c$, have been carried out in cells with a vertical height as small as 0.05 cm; the results are still affected by gravity within a reduced temperature of $|t/ \equiv |(T - T_c)/T_c| \le 10^{-4}$. The asymptotic region, where the leading singular term dominates the divergence of thermophysical properties, occurs in the same reduced temperature range. The ability to accurately experimentally test theoretical predictions has been severely limited because of the overlap of these two regions. It is for these reasons that MISTE was proposed to perform a set of thermodynamic measurements very close to the liquid-gas critical point of ^{3}He in a microgravity environment.

Theoretical Background

The distinguishing feature of critical phenomena associated with continuous phase transitions is the fact that thermophysical quantities exhibit power law behavior near critical points.[3] This power law behavior depends on the paths approaching the critical point as shown in Fig. 1 for the case of a liquid-gas critical point. For example, the isothermal susceptibility diverges as $\chi_T \propto \Gamma_0^+ t^{-\gamma}$ along the path of the critical isochore, $\rho = \rho_c$, above the transition and as $\chi_T \propto \Gamma_0^- |t|^{-\gamma'}$ along the coexistence curve below the transition. $\Gamma_0^\pm$ are the leading asymptotic critical amplitudes, with the superscript $\pm$ defining the single (+) or two-phase (−) regions. By contrast, the constant-volume heat capacity takes on the asymptotic power law form of $C_V \propto A_0^+ t^{-\alpha}$ along the critical isochore above T_c and $C_V \propto A_0^- |t|^{-\alpha'}$ below T_c. Here the

$$C_V^+ \to (A_0^+/\alpha)\, t^{-\alpha} \qquad \chi_T^+ \to \Gamma_0^+ t^{-\gamma}$$

Critical isochore

Critical isotherm $\quad (T_c, \rho_c) \quad \Delta P \to D\,\Delta\rho|\Delta\rho|^{\delta-1}$

$$C_V^- \to (A_0^-/\alpha)/|t|^{-\alpha'} \qquad \chi_T^- \to \Gamma_0^-/|t|^{-\gamma'}$$

Gas $\qquad\qquad\qquad\qquad$ Liquid

Coexistence curve

$\Delta\rho = \rho/\rho_c - 1$

$t = T/T_c - 1$
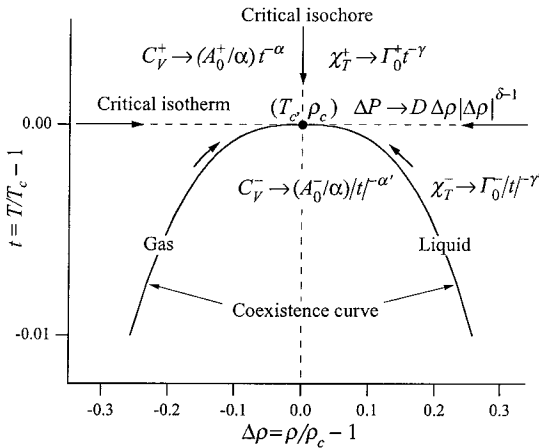
Fig. 1. Schematic representation of the liquid-gas critical region in the T-ρ plane.

quantities α α', γ, and γ' are known as critical exponents. Along the critical isotherm, $T = T_c$, the variation of pressure with density is given by $\Delta P \propto D\Delta\rho|\Delta\rho|^{\delta-1}$ where $\Delta P = (P - P_c)/P_c$ is the reduced pressure, $\Delta\rho = (\rho - \rho_c)/\rho_c$ is the reduced density and δ is the critical exponent that describes this singular behavior. In the two-phase region, the shape of the coexistence curve can be described as $\Delta\rho_{L,G} \equiv (\rho_{L,G} - \rho_c)/\rho_c \propto \pm B_0(-t)^\beta$, where β is a critical exponent, B_0 is the leading asymptotic critical amplitude, and L and G correspond to the liquid and gas phases respectively.

Scaling theories, using Renormalization Group (RG) theory, have calculated accurate values for the critical exponents and slightly less accurate values for critical amplitude ratios.[4,5] The scaling theories also predict scaling law relationships between the critical exponents that play an important role in asymptotic equations-of-state. An exact determination of the asymptotic region cannot be made theoretically because the leading critical amplitudes, such as $\Gamma_0^\pm$ or $A_0^\pm$, are system dependent and there are additional correction-to-scaling confluent singularities that also contain system-dependent amplitudes. Furthermore, most experimental measurements performed on the ground appear to be outside the asymptotic region. Because of these facts, the theoretical community has used RG theory to develop confluent singularity corrections to the asymptotic behavior. In real fluids, other non-asymptotic corrections due to liquid-gas asymmetry in the coexistence curve and analytic background terms also come into play.

In recent years there has been considerable interest in the behavior of fluid systems in the crossover region farther away from the transition.[6] This is the region where critical fluctuations no longer dominate the

system behavior and the system goes from critical behavior close to the transition described by scaling theories to mean field behavior farther away. Several crossover models have been proposed and need to be tested experimentally. All of these theoretical predictions require knowledge of the leading critical amplitudes that cannot be accurately determined from ground-based studies.

One of the main objectives of the MISTE flight experiment is to obtain gravity-free measurements at least two decades in reduced temperature closer to the transition along the critical isochore than can be obtained on the ground. Measurements will also be performed along a path of constant critical temperature, $T = T_c$, as a function of the sample density. These measurements should extend at least one decade in density closer to the transition. The planned microgravity experiments should provide sufficient additional measurements near the transition to obtain accurate asymptotic critical amplitudes for testing various asymptotic and crossover theoretical models.

Experimental Apparatus

In preparation for the MISTE flight experiment, measurements have been performed in a ground-based experimental cell that minimizes the effects of gravity. The sample volume is a flat cylindrical disk with a height of 0.05 cm and a diameter of 11.2 cm. Measurements of the susceptibility and heat capacity have been made in the gravity free region, $|t| > 10^{-4}$.

The ground-based cell temperature is measured using a magnetic susceptibility thermometer based on a SQUID magnetometer. This type of high-resolution thermometer (HRT) has a resolution of better than a nK (10^{-9} K). A miniaturized HRT, using a $GdCl_3$ paramagnetic salt, has been developed for studies at the ^{3}He critical point.[7] A calibrated Germanium resistance thermometer (GRT) is used to calibrate the HRT.

The fluid density is determined by measuring the ^{3}He dielectric constant using a parallel plate capacitor. The measured dielectric constant is then converted to density via the Clausius-Mossotti equation.[8]

A Straty-Adams type capacitive pressure gauge[9] has been fabricated to measure the in-situ sample pressure. The gauge uses a small portion of the cell boundary as a flexible diaphragm that moves in response to a pressure change.

A schematic of the flight cell is shown in Fig. 2. The flight cell will use two sets of temperature, density, and pressure sensors similar to what is being used on the ground. The candidate flight cell design is a cylindrical cell having dimensions of ~ 6 cm in diameter by ~ 4 cm in height that will provide ~ 8
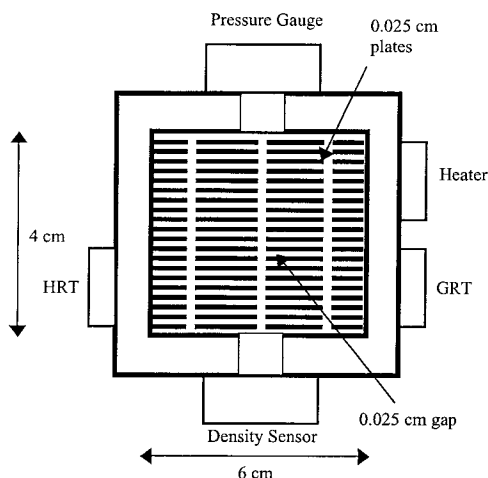
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Fig. 2. Schematic of the flight cell .

times the sample mass of the present ground-based cell to increase the accuracy of the heat capacity measurements. The flight cell will have 80 plates (0.025 cm thick) with holes in them and 80 spacers (0.025 cm thick) both made of copper and symmetrically situated in the cell. The fluid layers sandwiched between the plates will lead to an effective thermal time constant which will be ~ 4 times less than that obtained in the ground-based cell.

The flight experiment will also require a fluid transfer system to change the density in the cell. This system must allow the density to be changed between fixed values for measurements along isochores, and it must also be capable of slowly ramping the density for isotherm measurements. A schematic diagram of the proposed flight system is shown in Fig. 3. This system has a low-temperature valve that is closed during constant density measurements or open during density sweeps or other adjustments. With the valve open, the
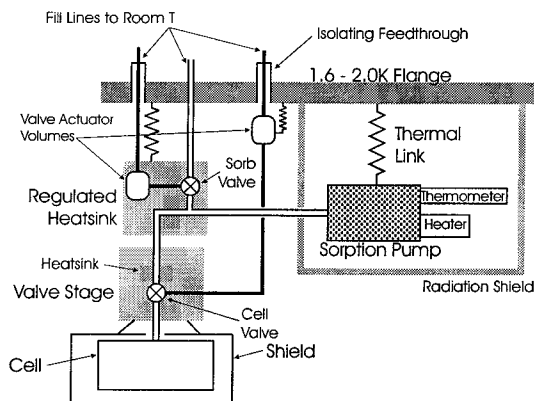

Fig. 3. Schematic of in-situ fluid transfer system.

amount of helium in the cell can be controlled over a wide range ($\pm$ 10%) by regulating the temperature of a sorption pump. A prototype system of this kind was used extensively in ground-based measurements and performed well.

The proposed flight in-situ system has one valve that will be remotely controllable on-orbit. This is the valve that isolates the cell from the sorption system for constant-volume heat capacity and electrostriction measurements. It will be opened and closed at least 25 times during the flight. Our baseline plan is to use a valve being developed in collaboration with Mission Research Corporation (MRC).[10] The valve leak rate requirement is < 10^{-6} std-cc helium/s. This leak rate is needed to maintain the density constant to 0.02% over a 1-month period, which is the maximum expected on-orbit time for runs at a given fixed density. The constant volume measurements with the most restrictive density requirements will be performed at the beginning of the data-taking period to minimize any adverse effects associated with a possible degradation of valve performance. The pressure to actuate the valve will be provided by a hot-volume system filled with helium.

Another valve will also be needed to isolate the low-temperature sample handling system from the fill line to room temperature. This valve would also be pressure actuated, but it would only need to be actuated a few times on the ground for initial sample filling.

Experimental Measurements

Measurements of the heat capacity and isothermal susceptibility have been performed in the MISTE ground-based cell. We have evaluated the drift and heat pulse methods for measuring the specific heat. The heat capacity is determined by knowing the amount of heat introduced into or extracted from the measurement cell and the resultant temperature change. Figure 4 shows heat capacity data close to the transition obtained from a continuous cooling drift run along the critical isochore. The data farther away from the transition (t > 2×10^{-4}) are consistent with earlier measurements.[11] In this study,[12] a radiation shield, surrounded the sample stage, was temperature controlled slightly below the critical temperature. The sample was initially regulated at a reduced temperature of ~ 3.5×10^{-4} above the transition. Then the regulation was removed and the sample was allowed to cool to the shield temperature. These data were taken with an average cooling drift rate of 3×10^{-4} K/hr. For this drift rate the overall density inhomogeneity in the sample due to the "piston effect",[13] which is in addition to the gravity stratification, is estimated to be $\leq 0.1\%$ in the single-
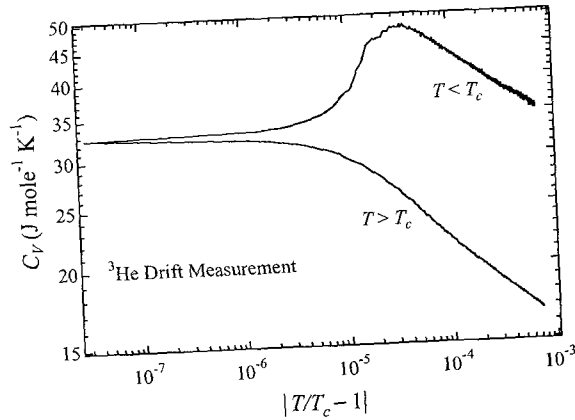
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Fig. 4. Cooling drift measurement of the heat capacity along the critical isochore.

phase region. These data clearly demonstrate the effects of gravity. Only measurements with reduced temperatures $|t| > 10^{-4}$ are essentially independent of gravity and can be used to test scaling predictions.

The isothermal susceptibility, $(\chi_T = \rho(\partial\rho/\partial P)_T)$, in the single-phase region, farther away from the transition $(t > 2\times10^{-4})$, was determined from PVT measurements. Figure 5 shows P-ρ measurements along several isotherms near the critical point in the range $-4.7\times10^{-3} < t < 3.0\times10^{-3}$. These measurements were performed by slowly removing fluid from the cell while holding the cell wall temperature constant. Fluid was removed from the cell by slowly reducing the temperature of the sorption pump, shown in Fig. 3, from a nominal value (~ 20 K). When the temperature of the sorption pump reached ~ 4 K, the sample density was approximately 25% below the critical value ($\Delta\rho \sim -0.25$). The cell could then be refilled to an initial density approximately 22% above the critical value ($\Delta\rho \sim 0.22$) by heating the sorption pump back to its nominal temperature. Measurements along a
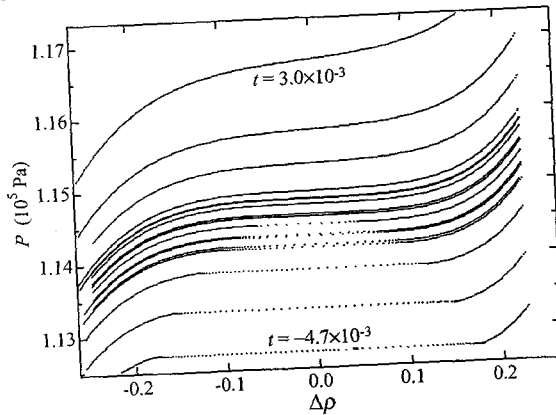
given isotherm typically took 5 – 10 hours. The isotherms near the critical temperature were measured at several different ramping rates to insure that quasi-equilibrium was attained.

For $T < T_c$, Fig. 5 clearly shows the phase boundary defining the coexistence curve. From the discontinuities in $(\partial P/\partial\rho)_T$, we can determine the coexistence curve shape. By including measurements obtained during the flight experiment, we should be able to determine the asymptotic and crossover critical amplitudes associated with the coexistence curve. In addition, by deriving $(\partial P/\partial\rho)_T$ in the single-phase region at the coexistence curve discontinuities, we can obtain the susceptibility along the coexistence curve, in the liquid and gas phases, from which the susceptibility critical amplitudes can be determined.

While the susceptibility χ_T along the critical isochore can be derived from PVT measurements, a pressure resolution of $\delta P/P \approx 1:10^{10}$ is required to measure χ_T with 1% uncertainty at $t \approx 10^{-6}$. This pressure resolution is very difficult to attain using conventional means. We have been evaluating a new electrostrictive technique that can attain this required pressure resolution.[14] This technique takes advantage of the fact that an electric field gradient can produce an equivalent pressure gradient within a dielectric fluid.[15] A field strength of 10^4 V/m is needed to generate a pressure gradient of $\delta P/P = 1:10^{10}$. In the case of a parallel plate capacitor with a gap of 150 μm, a DC voltage of only 1.6 V across the gap is needed to create this required electric field. The pressure difference between the inside and outside of the capacitor gap produces the density change that can be detected by a capacitance change. To a first approximation $\chi_T \propto \partial\rho/\partial E^2$.

Figure 6 illustrates the theoretically expected behavior E^2 versus $\Delta\rho$ within the capacitor gap along the isotherms $t = 1\times10^{-6}$ and $t = 0$ (critical isotherm). This calculation is for a capacitor gap $d = 150$ μm and assumes that the ambient cell density is 1% below the

Fig. 5. P-ρ measurements along isotherms near the critical point.

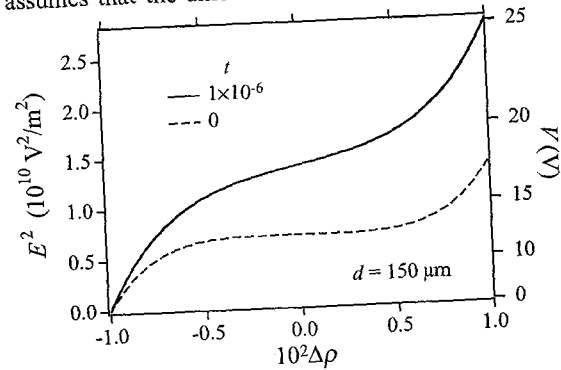

Fig. 6. Simulated square of electrical field versus induced density for $t = 1\times10^{-6}$ and $t = 0$.

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critical value. As the voltage across the capacitor is gradually increased, the density within the capacitor gap increases through the critical density. The susceptibility is determined from the inverse of the slope of the resultant curve.

Conclusion

We have provided an introduction to the MISTE flight experiment that will conduct a thorough investigation of the ^{3}He liquid-gas critical region. The thermodynamic measurements performed in the microgravity environment will be used to test asymptotic scaling theory predictions. The results of the microgravity asymptotic analysis will also permit a more stringent test of crossover models that describe behavior farther away from the transition.

Acknowledgements

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